Theory of correlated electron transport and inelastic tunneling spectroscopy

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(Dated: July 9, 2010)

For a non-superconducting system, the electronic tunneling current through an insulating barrier is calculated, including interaction effects. The exact Hamiltonian of the full system is projected onto the subspaces of the "left" and "right" leads. In the weak tunneling limit the well-known tunneling Hamiltonian is recovered, along with an additional term. This additional term originates from the projection of the electron-electron interaction onto each subsystem and corresponds to correlated tunneling. It is shown that the tunneling current is determined by—in addition to the single-particle density of states—the spin-spin and density-density susceptibilities. The signatures of which have recently been observed in several experiments.

PACS numbers: 73.40.Gk,72.10.-d

I. INTRODUCTION

Tunneling experiments were one of the first major confirmations of the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity.[1] These experiments showed a dramatic suppression in the conductance near the Fermi energy. The theoretical explanation of this was the formation of a gap in the quasi-particle density of states of the superconductor.[2] This essentially began the relationship linking a tunneling current in a many-body system to the single-particle density of states.

To describe tunneling at a general many-body level, Cohen *et al.*[3] later introduced the tunneling or transfer Hamiltonian, consisting of two disjoint, fully interacting Hamiltonians connected by a tunneling term. This tunneling term removes electrons from one system and creates them in the other. Although phenomenological, the tunneling Hamiltonian approach has become and remains the de facto standard for theoretically interpreting tunneling experiments in condensed matter, across a wide range of systems. Several attempts have been made to put many-body tunneling, with or without the tunneling Hamiltonian, on more rigorous theoretical ground,[4] but owing to the inherent difficulties of this problem, it still remains open.

Nevertheless, the standard tunneling formalism has been used extensively to account for an extraordinarily wide variety of experiments. Although, with the ever increasing refinement of experimental techniques, experiments are starting to probe physics not captured by this formalism. One area where modifications are needed is in the regime of inelastic tunneling, where the tunneling electron loses energy through interactions with the barrier or other additional degrees of freedom of the system being probed. These effects are typically accounted for by simply adding additional transfer terms.[5] Inelastic electron tunneling spectroscopy (IETS), most notably scanning tunneling microscopy IETS (STM-IETS), has be used to probe the internal vibrational modes of molecular adsorbates on surfaces. More recent experiments[6–8] on single magnetic atoms, finite spin chains, and magnetic substrates have shown evidence of spin related inelastic tunneling. Here, especially for the adatom chains, the current-voltage (I-V) curves show features where one would expect spin excitations of the system, such as the singlet-triplet excitation of neighboring pairs. Explaining such characteristics is outside the scope of the standard tunneling approach, which relates an I-V curve, or conductance, to single-particle properties, namely the local single-particle density of states. Spin excitations, such as the singlet-triplet transition, are manifestly two-particle properties, related to the spin (density) susceptibility or spin-spin correlation function. Several authors have been able to account for the experimental results of these experiments remarkably well,[9,10] by introducing an interaction effect between the STM tip and adatoms, leading to so-called spin mediated tunneling. The microscopic origin of such a term and how it should be generalized, to say other spin excitations such as magons,[8] isn't immediately clear though. In principle these and other interaction effects should occur in virtually all tunneling experiments.[11] One would like a theoretical foundation that is first principles driven such that it could be applied to a wide variety of systems and also be generalizable to others in a systematic manner. The main goal of this article is to give such a derivation of the tunneling current, in the spirit of the transfer Hamiltonian, that includes these interaction or two-body effects.

In the next section we start with a general interacting Hamiltonian for a system with a tunneling barrier. The original Hamiltonian is then projected onto the low-energy (below the height of the barrier) "left" and "right" subspaces. This projection is conjectured to capture the most salient tunneling processes. From which, the standard tunneling Hamiltonian is recovered along with an additional term, which describes correlated or interaction meditated tunneling, stemming from the original electron-electron interaction. In Sec. III we calculate the steady-state tunneling current through the barrier with respect to this effective tunneling Hamiltonian. The result gives an additional contribution to the current from two-particle susceptibilities, as well as the well-known contribution determined by the single-particle density of states.

II. DERIVATION OF EFFECTIVE TUNNELING HAMILTONIAN

Here we consider a one-dimensional system for notational convenience; the generalization to higher dimensions is straightforward. For an interacting system in the presence of a tunneling barrier, the grand-canonical Hamiltonian of the *entire* system is given in second quantization by (setting $\hbar = 1$)

$$H = \sum_{\sigma} \int dx\, \Psi^{\dagger}_{\sigma}(x) \Big[-\frac{\nabla^2}{2m} - \mu + V(x) \Big] \Psi_{\sigma}(x) + \frac{1}{2} \sum_{\sigma,\sigma'} \int dx dx'\, \Psi^{\dagger}_{\sigma}(x) \Psi^{\dagger}_{\sigma'}(x') U(x,x') \Psi_{\sigma'}(x') \Psi_{\sigma}(x), \tag{1}$$

where $U(x,x')$ is the electron-electron interaction (assumed to be symmetric), μ is the chemical potential, and $V(x)$ is taken to be the tunneling barrier, but could also contain disorder, lattice fields or other single-particle potentials, see Fig. 1. The field operators $\Psi(x)$ and $\Psi^{\dagger}(x)$ obey the standard fermionic anticommuation relations $\{\Psi_{\sigma}(x), \Psi_{\sigma'}(x')\} = 0$, $\{\Psi_{\sigma}(x), \Psi^{\dagger}_{\sigma'}(x')\} = \delta_{\sigma,\sigma'}\delta(x-x')$, and can be expressed in terms of mode creation and annihilation operators by

$$\Psi_{\sigma}(x) = \sum_{k} \psi_{k\sigma}(x) a_{k\sigma} \tag{2}$$

$$\Psi^{\dagger}_{\sigma}(x) = \sum_{k} \psi^{*}_{k\sigma}(x) a^{\dagger}_{k\sigma}, \tag{3}$$

where $\{a_{k\sigma}, a_{k'\sigma'}\} = 0$, $\{a_{k\sigma}, a^{\dagger}_{k'\sigma'}\} = \delta_{\sigma,\sigma'}\delta_{k,k'}$, and $\psi_{k\sigma}(x)$ are the exact single-particle eigenstates of (1).

Next, we consider two related Hamiltonians

$$H_{\rm L} = \sum_{\sigma} \int dx\, \Psi^{\dagger}_{{\rm L},\sigma}(x) \Big[-\frac{\nabla^2}{2m} - \mu + V_{\rm L}(x) \Big] \Psi_{{\rm L},\sigma}(x) + \frac{1}{2} \sum_{\sigma,\sigma'} \int dx dx'\, \Psi^{\dagger}_{{\rm L},\sigma}(x) \Psi^{\dagger}_{{\rm L},\sigma'}(x') U(x,x') \Psi_{{\rm L},\sigma'}(x') \Psi_{{\rm L},\sigma}(x), \tag{4}$$

and

$$H_{\rm R} = \sum_{\sigma} \int dx\, \Psi^{\dagger}_{{\rm R},\sigma}(x) \Big[-\frac{\nabla^2}{2m} - \mu + V_{\rm R}(x) \Big] \Psi_{{\rm R},\sigma}(x) + \frac{1}{2} \sum_{\sigma,\sigma'} \int dx dx'\, \Psi^{\dagger}_{{\rm R},\sigma}(x) \Psi^{\dagger}_{{\rm R},\sigma'}(x') U(x,x') \Psi_{{\rm R},\sigma'}(x') \Psi_{{\rm R},\sigma}(x), \tag{5}$$

where

$$V_{\rm L}(x) = \begin{cases} V(x), & \text{for } x \le a \\ V(a), & \text{for } x \ge a\,, \end{cases} \tag{6}$$

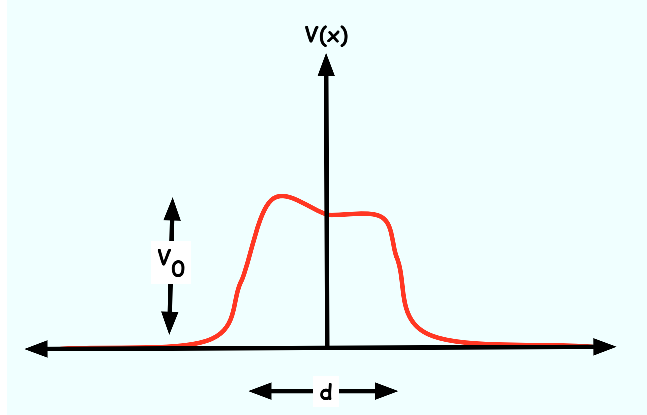

FIG. 1. Schematic of a tunneling barrier for the full Hamiltonian, Eq. (1), having a characteristic width d and height V_0.

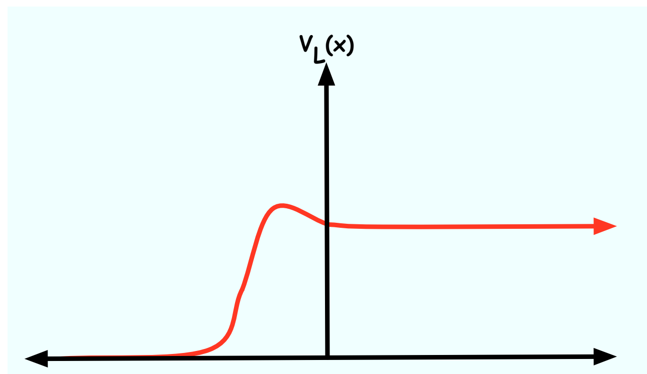

FIG. 2. Potential barrier for $H_{\rm L}$, defined in (6).

and

$$V_{\rm R}(x) = \begin{cases} V(x), & \text{for } x \ge a \\ V(a), & \text{for } x \le a. \end{cases} \tag{7}$$

The parameter a is an arbitrary point within the barrier, such that $V(x) = V_{\rm L}(x)\Theta(a-x) + V_{\rm R}(x)\Theta(x-a)$, where $\Theta(x)$ is the Heavyside step function; see Figs. 2 and 3. The field operators in (4) and (5) are given by

$$\Psi_{{\rm L},\sigma}(x) = \sum_{k} \psi_{{\rm L},k\sigma}(x) a_{{\rm L},k\sigma} \tag{8}$$

$$\Psi_{{\rm R},\sigma}(x) = \sum_{k} \psi_{{\rm R},k\sigma}(x) a_{{\rm R},k\sigma}, \tag{9}$$

where $\psi_{{\rm L},k\sigma}$ and $\psi_{{\rm R},k\sigma}$ are the exact and complete single-particle eigenstates of $H_{\rm L}$ and $H_{\rm R}$ respectively.

The Hilbert space of H, $\mathcal{H}$ can be decomposed into the Hilbert spaces of $H_{\rm L}$ and $H_{\rm R}$,

$$\mathcal{H} = \mathcal{H}_{\rm L} + \mathcal{H}_{\rm R} + \delta\mathcal{H}, \tag{10}$$

where $\delta\mathcal{H}$ allows for states outside of either $\mathcal{H}_{\rm L}$ or $\mathcal{H}_{\rm R}$, e.g. resonant states within the barrier itself. Such possibilities will not be considered here. One can then define

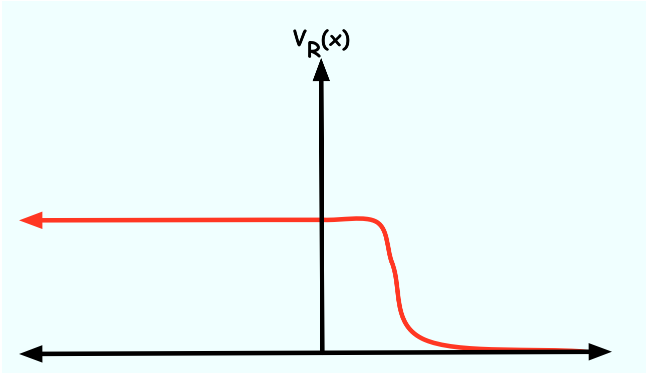

FIG. 3. Potential barrier for $H_{\rm R}$, defined in (7).

projection operators

$$P_{\rm L} = \sum_\alpha |\psi_{{\rm L},\alpha}\rangle\langle\psi_{{\rm L},\alpha}|$$
$$P_{\rm R} = \sum_\alpha |\psi_{{\rm R},\alpha}\rangle\langle\psi_{{\rm R},\alpha}|, \tag{11}$$

which project the original Hilbert space on to the left and right subspaces respectively. We assume that the sum of projection operators, to a good approximation, forms a resolution of the identity in $\mathcal{H}$, i.e.

$$P_{\rm L} + P_{\rm R} \simeq \mathbb{1}. \tag{12}$$

The electron field operator of (1) can therefore be written as

$$\Psi_\sigma(x) = (P_{\rm L}^\dagger + P_{\rm R}^\dagger)\Psi_\sigma(x)(P_{\rm L} + P_{\rm R})$$
$$= \tilde{\Psi}_{\rm L}(x) + \tilde{\Psi}_{\rm R}(x) + \delta\Psi(x), \tag{13}$$

where $\delta\Psi(x)$ is of order of the tunneling amplitude, i.e., for states below the barrier height $\delta\Psi(x) \sim O(T_{\alpha,\alpha'} = \langle\psi_{{\rm L},\alpha}|V|\psi_{{\rm R},\alpha'}\rangle)$. $\delta\Psi(x)$ will be neglected from here on, as it will ultimately correspond to higher order terms, in the tunneling amplitude, in a calculation of the current. Projecting the field operator of the original Hamiltonian onto the left and right subspaces

$$\tilde{\Psi}_{{\rm L},\sigma}(x) = P_{\rm L}^\dagger\Psi_\sigma(x)P_{\rm L} = \sum_k \psi_{k\sigma}(x)a_{{\rm L},{\rm k}\sigma}$$
$$\tilde{\Psi}_{{\rm L},\sigma}^\dagger(x) = P_{\rm L}^\dagger\Psi_\sigma^\dagger(x)P_{\rm L} = \sum_k \psi_{k\sigma}^*(x)a_{{\rm L},{\rm k}\sigma}^\dagger, \tag{14}$$

and

$$\tilde{\Psi}_{{\rm R},\sigma}(x) = P_{\rm R}^\dagger\Psi_\sigma(x)P_{\rm R} = \sum_k \psi_{k\sigma}(x)a_{{\rm R},{\rm k}\sigma}$$
$$\tilde{\Psi}_{{\rm R},\sigma}^\dagger(x) = P_{\rm R}^\dagger\Psi_\sigma^\dagger(x)P_{\rm R} = \sum_k \psi_{k\sigma}^*(x)a_{{\rm R},{\rm k}\sigma}^\dagger, \tag{15}$$

where $a_{{\rm L},{\rm k}\sigma} = \int dx\,\psi_{{\rm L},k\sigma}(x)\Psi_\sigma(x)$, $a_{{\rm L},{\rm k}\sigma}^\dagger = \int dx\,\psi_{{\rm L},k\sigma}^*(x)\Psi_\sigma^\dagger(x)$, $a_{{\rm R},{\rm k}\sigma} = \int dx\,\psi_{{\rm R},k\sigma}(x)\Psi_\sigma(x)$, and $a_{{\rm R},{\rm k}\sigma}^\dagger = \int dx\,\psi_{{\rm R},k\sigma}^*(x)\Psi_\sigma^\dagger(x)$. From which it readily follows that $\{a_{{\rm L},{\rm k}\sigma}, a_{{\rm L},{\rm k}'\sigma'}^\dagger\} = \{a_{{\rm R},{\rm k}\sigma}, a_{{\rm R},{\rm k}'\sigma'}^\dagger\} = \delta_{\sigma,\sigma'}\delta_{k,k'}$, $\{a_{{\rm L},{\rm k}\sigma}, a_{{\rm L},{\rm k}'\sigma'}\} = \{a_{{\rm R},{\rm k}\sigma}, a_{{\rm R},{\rm k}'\sigma'}\} = \{a_{{\rm L},{\rm k}\sigma}, a_{{\rm R},{\rm k}'\sigma'}\} = 0$, and $\{a_{{\rm L},{\rm k}\sigma}, a_{{\rm R},{\rm k}'\sigma'}^\dagger\} = \langle\psi_{{\rm L},k\sigma}|\psi_{{\rm R},k'\sigma'}\rangle$. In general the left and right states are not orthogonal; $\langle\psi_{{\rm L},\alpha}|\psi_{{\rm R},\alpha'}\rangle \neq 0$. For states with energy below the barrier height $\langle\psi_{{\rm L},\alpha}|\psi_{{\rm R},\alpha'}\rangle \sim O(T_{\alpha,\alpha'})$.

As the exact single-particle eigenstates of (1), (4), and (5) are assumed to form a complete basies, we can expand the original eigenfunctions in terms of the left and right basis

$$\psi_{k\sigma}(x) = \sum_{k'} c_{k,k'}^{\rm L}\psi_{{\rm L},k'\sigma}(x) \tag{16}$$

$$\psi_{k\sigma}(x) = \sum_{k'} c_{k,k'}^{\rm R}\psi_{{\rm R},k'\sigma}(x), \tag{17}$$

where $c_{k,k'}^{\rm L} = \langle\psi_{{\rm L},k}|\psi_{k'}\rangle$ and $c_{k,k'}^{\rm R} = \langle\psi_{{\rm R},k}|\psi_{k'}\rangle$. The expansion coefficients can be obtained from assuming an explicit form of the tunneling barrier[4] or within WKB (Wentzel-Kramers-Brillouin) theory for an arbitrary potential.[12] For a wide tall barrier and for states with energy much less than the barrier height $c_{k,k'}^{\rm L} \propto \delta_{k,k'} + O(T_{k,k'})$ and $c_{k,k'}^{\rm R} \propto \delta_{k,k'} + O(T_{k,k'})$. Thus, apart from an overall proportionality constant, to leading order in the tunneling amplitude

$$\tilde{\Psi}_{{\rm L},\sigma}(x) \simeq \Psi_{{\rm L},\sigma}(x) + O(T_{k,k'}) \tag{18}$$

$$\tilde{\Psi}_{{\rm R},\sigma}(x) \simeq \Psi_{{\rm R},\sigma}(x) + O(T_{k,k'}). \tag{19}$$

Therefore, the original fields operators, to leading order in the tunneling, can be decomposed into the left and right subsystems by

$$\Psi_\sigma(x) \simeq \Psi_{{\rm L},\sigma}(x) + \Psi_{{\rm R},\sigma}(x) + O(T_{k,k'})$$
$$\Psi_\sigma^\dagger(x) \simeq \Psi_{{\rm L},\sigma}^\dagger(x) + \Psi_{{\rm R},\sigma}^\dagger(x) + O(T_{k,k'}). \tag{20}$$

It should be noted that, at this point, because we have restricted ourselves to states below the barrier, the field operators $\Psi_{\rm L}^\dagger(x)$ and $\Psi_{\rm R}^\dagger(x)$ in Eq. (20), no longer create (annihilate) electrons in localized delta-function states, but instead in some broaden approximation. We will not deal with this technicality and assume that for a high barrier, compared to the Fermi energy, to a good approximation the states below the barrier form a sufficiently complete set.

Expressing the exact Hamiltonian (1) in terms of the approximate projected operators (20), and for now neglecting interactions, gives

$$H \approx \sum_{k,\sigma}(\epsilon_{k\sigma} - \mu)a_{{\rm L},{\rm k}\sigma}^\dagger a_{{\rm L},{\rm k}\sigma} + \sum_{k,\sigma}(\epsilon_{k\sigma} - \mu)a_{{\rm R},{\rm k}\sigma}^\dagger a_{{\rm R},{\rm k}\sigma}$$
$$+ \sum_{k,k',\sigma}\left[T_{k,k'}a_{{\rm L},{\rm k}\sigma}^\dagger a_{{\rm R},{\rm k}'\sigma} + {\rm H.c.}\right]$$
$$+ \sum_{k,k',\sigma}\left[\xi_{k,k',\sigma}(\epsilon_{k'\sigma} - \mu)a_{{\rm L},{\rm k}\sigma}^\dagger a_{{\rm R},{\rm k}'\sigma} + {\rm H.c.}\right], \tag{21}$$

where terms such as $\int dx\, \Psi^\dagger_{\rm L}(x)V(x)\Psi_{\rm L}(x)$ that only lead to a change in the density and energies near the barrier have been neglected, and $\xi_{k,k'} = \langle \psi_{{\rm L},k}|\psi_{{\rm R},k'}\rangle$. At this level, the fact that the eigenstates of the left and right sides are not orthogonal simply leads to a renormalization of the tunneling matrix elements, $T_{k,k'} \to T_{k,k'} + \xi_{k,k'}(\epsilon_{k'} - \mu)$. Even in the approximation $\xi_{k,k'} \to 0$, the important and dominate physics is stilled captured. Thus, neglecting these terms or equivalently setting $\xi_{k,k'} = 0$, one can still expect to obtain quantitatively correct results, of course this approximation can be relaxed.[13] Also, this simplification exactly reduces (21) to the standard tunneling Hamiltonian. In addition, this greatly simplifies calculations of expectation values, i.e Green's functions, as now $\{\Psi_{{\rm L},\sigma}(x), \Psi^\dagger_{{\rm R},\sigma}(x)\} = 0$, which we will assume from here on. With such an approximation we have essentially arrived back at the starting point for the standard tunneling Hamiltonian. One may wonder why all of the previous formalities were necessary. For one, it demonstrates what approximations and assumptions are made when one is using the standard tunneling Hamiltonian formalism, and secondly it will allow us to treat the effects of interactions on tunneling quantitatively, which we now turn to.

Normally, interactions are introduced simply by adding the appropriate interactions terms to the left and right subsystems. This neglects the cross terms generated by expressing the original operators in terms of the approximate left and right states. As we will see, it is these cross terms that are responsible for the electronic inelastic tunneling properties. Expressing the interaction term of (1) in terms of the projected operators, Eqs. (20), leads to (see the Appendix A for details)

$$H \approx H_{\rm L} + H_{\rm R} + H_{\rm T}, \tag{22}$$

where $H_{\rm L}$, $H_{\rm R}$ are the fully interacting Hamiltonians of the left and right subsystems

$$\begin{aligned} H_{\rm L} &= \sum_\sigma \int dx\, \Psi^\dagger_{{\rm L},\sigma}(x)\Big[-\frac{\nabla^2}{2m} - \mu\Big]\Psi_{{\rm L},\sigma}(x) + \frac{1}{2}\sum_{\sigma,\sigma'}\int dxdx'\, \Psi^\dagger_{{\rm L},\sigma}(x)\Psi^\dagger_{{\rm L},\sigma'}(x')U(x,x')\Psi_{{\rm L},\sigma'}(x')\Psi_{{\rm L},\sigma}(x), \\ H_{\rm R} &= \sum_\sigma \int dx\, \Psi^\dagger_{{\rm R},\sigma}(x)\Big[-\frac{\nabla^2}{2m} - \mu\Big]\Psi_{{\rm R},\sigma}(x) + \frac{1}{2}\sum_{\sigma,\sigma'}\int dxdx'\, \Psi^\dagger_{{\rm R},\sigma}(x)\Psi^\dagger_{{\rm R},\sigma'}(x')U(x,x')\Psi_{{\rm R},\sigma'}(x')\Psi_{{\rm R},\sigma}(x), \end{aligned} \tag{23}$$

and the tunneling or transfer part is given by

$$H_{\rm T} = \sum_\sigma \int dx\, \big[T(x)\Psi^\dagger_{{\rm L},\sigma}(x)\Psi_{{\rm R},\sigma}(x) + {\rm H.c.}\big] + \sum_\sigma \int dxdx'\, U(x,x')\Big\{\Psi^\dagger_{{\rm L},\sigma}(x)\Psi_{{\rm R},\sigma}(x)\big[\hat{n}_{\rm L}(x') + \hat{n}_{\rm R}(x')\big] + {\rm H.c}\Big\}, \tag{24}$$

where $\hat{n}(x) = \sum_\sigma \Psi^\dagger_\sigma(x)\Psi_\sigma(x)$. The first term of (24) is the standard single-particle tunneling, which comes from the kinetic energy term of the original Hamiltonian. The last term is from the interaction of the original system and is normally not considered in most treatments of tunneling. This term involves a interaction mediated transfer of electrons, or correlated hopping in a lattice model.[14] Although, this term might be small, as it involves the correlated transfer electrons, it is ultimately responsible for the inelastic tunneling properties.

III. EVALUATION OF THE TUNNELING CURRENT

In the presence of an applied electric field a net current will flow through the barrier. Here we will calculate the steady-state current in the limit of weak tunneling between the left and right sub-systems. As usual,[15] we will assume that each subsystem is separately in thermodynamic equilibrium, where the chemical potential of each side differs only by the applied voltage eV. The drop in the chemical potential is assumed to occur entirely within the barrier. Because the total particle number commutes with the effective Hamiltonian, the current is defined to be proportional to the expectation value of the time-rate-change of the number of electrons in the left (or right) sub-system. By Heisenberg's equation of motion ($e > 0$) the current operator is given by

$$\hat{I} = -e\partial_t \hat{N}_{\rm L} = -ie\big[\hat{N}_{\rm L}, H\big] = -ie\big[\hat{N}_{\rm L}, H_{\rm T}\big], \tag{25}$$

where $\hat{N}_{\rm L} = \sum_\sigma \int dx\, \Psi^\dagger_{{\rm L}\sigma}(x)\Psi_{{\rm L}\sigma}(x)$. Evaluating the commutator gives

$$\hat{I} = -ie\sum_{\sigma}\int dx\,\left[T(x)\Psi^{\dagger}_{\mathrm{L},\sigma}(x)\Psi_{\mathrm{R},\sigma}(x) - \mathrm{H.c.}\right] - ie\sum_{\sigma}\int dxdx'\,U(x,x')\Big\{\Psi^{\dagger}_{\mathrm{L},\sigma}(x)\Psi_{\mathrm{R},\sigma}(x)\left[\hat{n}_{\mathrm{L}}(x') + \hat{n}_{\mathrm{R}}(x')\right] - \mathrm{H.c}\Big\}. \tag{26}$$

The nonequilibrium expectation value of (26) gives the measured tunneling current. Here we obtain this expectation within linear response theory, treating the tunneling term, H_{T}, as the perturbation. To leading order in the tunneling, the current is given by the thermal expectation of

$$I = i\int_{-\infty}^{t} dt'\left\langle\left[H_{\mathrm{T}}(t'), \hat{I}(t)\right]\right\rangle_{H_0}, \tag{27}$$

where $\hat{O}(t) = e^{iH_0t}\hat{O}e^{-iH_0t}$, $H_0 = H_{\mathrm{L}} + H_{\mathrm{R}}$, and we have assumed that as $t \to -\infty$ the two sub-systems are completely decoupled, i.e. the height of the barrier is infinitely large.

Evaluating (27) for a non-superconducting system and assuming local tunneling, i.e., the wave functions of one side are spatially localized about a point x, as is the case for an STM, the total current can be written as (See Appendix B for a complete derivation.)

$$I = I_1 + I_2. \tag{28}$$

The first term gives the standard expression for the tunneling current.

$$I_1 = 2\pi e|T|^2\sum_{\sigma}\int d\omega\,\rho_{\mathrm{L},\sigma}(x,\omega+eV)\rho_{\mathrm{R},\sigma}(x,\omega) \times\left[n_{\mathrm{F}}(\omega) - n_{\mathrm{F}}(\omega+eV)\right], \tag{29}$$

which relates the single-particle local density of states

$$\rho_{\sigma}(x,\omega) = -\frac{1}{\pi}\mathrm{Im}\,G^{\mathrm{ret}}_{\sigma}(x,\omega), \tag{30}$$

to the total current, where G^{ret} is the retarded single-particle Green's function and $n_{\mathrm{F}}(\omega) = (\exp(\beta\omega)+1)^{-1}$. The second term is related to the electronic inelastic tunneling and is in general given by

$$\begin{aligned}
I_2 =\;& \\
& eU^2\sum_{\sigma,\sigma'}\int_{-\infty}^{\infty} d\omega d\omega'\,\rho_{\mathrm{L},\sigma}(x,\omega+eV)\rho_{\mathrm{R},\sigma'}(x,\omega')\Big\{\left[\chi^{\mathrm{L}}_{\overline{\sigma}',\overline{\sigma},\overline{\sigma},\overline{\sigma}'}(x,\omega'-\omega-eV) + \chi^{\mathrm{R}}_{\overline{\sigma}',\overline{\sigma},\overline{\sigma},\overline{\sigma}'}(x,\omega'-\omega)\right]n_{\mathrm{F}}(\omega')\left[1-n_{\mathrm{F}}(\omega+eV)\right] \\
& - \left[\chi^{\mathrm{L}}_{\overline{\sigma},\overline{\sigma}',\overline{\sigma}',\overline{\sigma}}(x,\omega-\omega'+eV) + \chi^{\mathrm{R}}_{\overline{\sigma},\overline{\sigma}',\overline{\sigma}',\overline{\sigma}}(x,\omega-\omega')\right]n_{\mathrm{F}}(\omega+eV)\left[1-n_{\mathrm{F}}(\omega')\right]\Big\} \\
& + e\,\pi U^2\sum_{\sigma}\int_{-\infty}^{\infty} d\omega d\omega'\,\rho_{\mathrm{L},\overline{\sigma}}(x,\omega+eV)\rho_{\mathrm{R},\sigma}(x,\omega')\rho^{\mathrm{II}}_{\mathrm{L},\sigma,\overline{\sigma}}(x,\omega+\omega'+eV) \\
& \times\Big\{n_{\mathrm{F}}(\omega+eV)n_{\mathrm{F}}(\omega')\left[1-n_{\mathrm{F}}(\omega+\omega'+eV)\right] - \left[1-n_{\mathrm{F}}(\omega+eV)\right]\left[1-n_{\mathrm{F}}(\omega')\right]n_{\mathrm{F}}(\omega+\omega'+eV)\Big\} \\
& - e\,\pi U^2\sum_{\sigma}\int_{-\infty}^{\infty} d\omega d\omega'\,\rho_{\mathrm{L},\overline{\sigma}}(x,\omega+eV)\rho_{\mathrm{R},\sigma}(x,\omega')\rho^{\mathrm{II}}_{\mathrm{R},\sigma,\overline{\sigma}}(x,\omega+\omega') \\
& \times\Big\{n_{\mathrm{F}}(\omega+eV)n_{\mathrm{F}}(\omega')\left[1-n_{\mathrm{F}}(\omega+\omega')\right] - \left[1-n_{\mathrm{F}}(\omega+eV)\right]\left[1-n_{\mathrm{F}}(\omega')\right]n_{\mathrm{F}}(\omega+\omega')\Big\},
\end{aligned} \tag{31}$$

where $\chi^{X}_{\sigma_1,\sigma_2,\sigma_3,\sigma_4}(x,t) = \left\langle\Psi^{\dagger}_{X,\sigma_1}(x,t)\Psi_{X,\sigma_2}(x,t)\Psi^{\dagger}_{X,\sigma_3}(x,0)\Psi_{X,\sigma_4}(x,0)\right\rangle_{H_X}$ and $\rho^{\mathrm{II}}_{\sigma,\overline{\sigma}}(x,\omega) = -\frac{1}{\pi}\mathrm{Im}\,G^{\mathrm{II}}_{\sigma,\overline{\sigma}}(x,\omega)$ is the two-particle density of states, with

$$G^{\mathrm{II}}_{\sigma,\overline{\sigma}}(x,t) = -i\theta(t)\langle\{\Psi_{\sigma}(x,t)\Psi_{\overline{\sigma}}(x,t), \Psi^{\dagger}_{\overline{\sigma}}(0)\Psi^{\dagger}_{\sigma}(0)\}\rangle_{H}, \tag{32}$$

and $\bar{\sigma}$ is the opposite of σ. Note that χ is not a time-ordered expectation, but is instead, apart from thermal factors, related to a two-particle spectral function. For a system with weak or no correlations on one side and a spin-independent featureless density of states near the Fermi energy ϵ_{F} and neglecting the contributions from the two-particle density of states,

$$I_2 \approx eU^2\rho_{\mathrm{L}}(\epsilon_{\mathrm{F}})\rho_{\mathrm{R}}(\epsilon_{\mathrm{F}})\sum_{\sigma,\sigma'}\int_{-\infty}^{\infty} d\omega d\omega'\,\chi^{\mathrm{R}}_{\sigma',\sigma,\sigma,\sigma'}(x,\omega-\omega')\Big\{n_{\mathrm{F}}(\omega)\left[1-n_{\mathrm{F}}(\omega'+eV)\right] - n_{\mathrm{F}}(\omega+eV)\left[1-n_{\mathrm{F}}(\omega')\right]\Big\}. \tag{33}$$

Equation (33) can be rewritten using

$$\sum_{\sigma,\sigma'} \chi^{\mathrm{R}}_{\sigma',\sigma,\sigma,\sigma'}(x,t) = 2S_{\mathrm{R}}(x,t) + \frac{1}{2}\Pi_{\mathrm{R}}(x,t)$$
$$= 2\langle \mathbf{s}_{\mathrm{R}}(x,t)\cdot\mathbf{s}_{\mathrm{R}}(x,0)\rangle + \frac{1}{2}\langle \hat{n}_{\mathrm{R}}(x,t)\hat{n}_{\mathrm{R}}(x,0)\rangle, \tag{34}$$

where $\langle \mathbf{s}_{\mathrm{R}}(x,t)\cdot\mathbf{s}_{\mathrm{R}}(x,0)\rangle$ and $\langle \hat{n}_{\mathrm{R}}(x,t)\hat{n}_{\mathrm{R}}(x,0)\rangle$ are the spin-spin (density) and density-density correlation functions respectively, giving

$$I_2 = 2eU^2\rho_{\mathrm{L}}(\epsilon_{\mathrm{F}})\rho_{\mathrm{R}}(\epsilon_{\mathrm{F}}) \int_{-\infty}^{\infty} d\omega d\omega'\, S_{\mathrm{R}}(x,\omega-\omega')\Big\{n_{\mathrm{F}}(\omega)\big[1-n_{\mathrm{F}}(\omega'+eV)\big] - n_{\mathrm{F}}(\omega+eV)\big[1-n_{\mathrm{F}}(\omega')\big]\Big\}$$
$$+\frac{eU^2\rho_{\mathrm{L}}(\epsilon_{\mathrm{F}})\rho_{\mathrm{R}}(\epsilon_{\mathrm{F}})}{2} \int_{-\infty}^{\infty} d\omega d\omega'\, \Pi_{\mathrm{R}}(x,\omega-\omega')\Big\{n_{\mathrm{F}}(\omega)\big[1-n_{\mathrm{F}}(\omega'+eV)\big] - n_{\mathrm{F}}(\omega+eV)\big[1-n_{\mathrm{F}}(\omega')\big]\Big\}. \tag{35}$$

For spin systems, such as those of Refs. [6–8], the density fluctuation term can be safely neglected. This reduces Eq. (35) to the form of those proprosed in Refs. 9 and 10, which have accurately described the experiments of Refs. 6 and 7. But in general, Eq. (31) gives the total correlated transfer contribution to the tunneling current and can be used in a broader range of experiments, which will be the subject of a future publications.

Within the approximations leading to Eq. (35) and in the low temperature limit, $k_{\mathrm{B}}T \ll \epsilon_{\mathrm{F}}$, the differential conductance $G = \partial_V I$ is

$$G(eV) = 4\pi e^2|T|^2\rho_{\mathrm{L}}(\epsilon_{\mathrm{F}})\rho_{\mathrm{R}}(\epsilon_{\mathrm{F}}) + 2e^2U^2\rho_{\mathrm{L}}(\epsilon_{\mathrm{F}})\rho_{\mathrm{R}}(\epsilon_{\mathrm{F}})$$
$$\times\Bigg\{\int d\omega\, S_{\mathrm{R}}(x,\omega)\big[n_{\mathrm{F}}(\omega+eV)+n_{\mathrm{F}}(\omega-eV)\big]$$
$$+\frac{1}{4}\int d\omega\, \Pi_{\mathrm{R}}(x,\omega)\big[n_{\mathrm{F}}(\omega+eV)+n_{\mathrm{F}}(\omega-eV)\big]\Bigg\}. \tag{36}$$

The second derivative of the current gives

$$\partial_V^2 I = 2e^3U^2\rho_{\mathrm{L}}(\epsilon_{\mathrm{F}})\rho_{\mathrm{R}}(\epsilon_{\mathrm{F}})\Big\{S_{\mathrm{R}}(x,eV) - S_{\mathrm{R}}(x,-eV)$$
$$+\frac{1}{4}\big[\Pi_{\mathrm{R}}(x,eV) - \Pi_{\mathrm{R}}(x,-eV)\big]\Big\}. \tag{37}$$

IV. OUTLOOK

As mentioned in the introduction and demonstrated in the body of the article, in principle all tunneling experiments, at some level, probe two-particle properties induced by interactions effects. Although, by their very nature these effects can be small, when compared to the single-particle terms. It takes a well engineered experiment, such as those of Refs. [6–8], to separate the contributions. Another system where such effects have probably been observed, is in transport through a quantum point contact (QPC). It is believed that interactions are responsible for the "0.7 conductance anomaly" of QPCs.[16] A lot of theoretical work has gone into explaining this effect. For instance, in Ref. [17] a generalized Anderson model, which effectively contains correlated transfer of electrons, has been applied with some success to this problem. In the previous sections we have calculated the contribution from interactions to the many-body tunneling current within an "STM-like" geometry. A natural extension would be to apply the ideas developed here to a quantum-dot geometry or QPC. Such a derivation would lead to a generalization of the well-known Meir-Wingreen expression.[18]

Furthermore, one could use the results presented here to design experiments to probe the two-particle properties of other important and interesting systems. For example, how these properties change as one approaches the superconducting instability. This could give information about the pairing mechanism of high-temperature superconductors.

ACKNOWLEDGMENTS

The author would like to thank Michael Geller, Hartmut Hafermann, Mikhail Katsnelson, and Alexander Lichtenstein for many useful conversations. This work has been supported by the German Research Council (DFG) under SFB 668 and the Louisiana Board of Regents.

Appendix A: Details of the derivation of the tunneling Hamiltonian

Here we give the details of the derivation of Eq. (22). The kinetic energy term was done in the body of the paper, here we will deal with the interaction term. Expressing the interaction term of (1) in terms of the projected operators, Eqs. (20) and keeping only terms to leading

order in the tunneling amplitude gives

$$
\begin{aligned}
&\frac{1}{2}\sum_{\sigma,\sigma'}\int dxdx'\,\Psi^\dagger_\sigma(x)\Psi^\dagger_{\sigma'}(x')U(x,x')\Psi_{\sigma'}(x')\Psi_\sigma(x) = \\
&\underbrace{\frac{1}{2}\sum_{\sigma,\sigma'}\int dxdx'\,\Psi^\dagger_{\mathrm{L},\sigma}(x)\Psi^\dagger_{\mathrm{L},\sigma'}(x')U(x,x')\Psi_{\mathrm{L},\sigma'}(x')\Psi_{\mathrm{L},\sigma}(x)}_{\text{Left interaction}} + \underbrace{\frac{1}{2}\sum_{\sigma,\sigma'}\int dxdx'\,\Psi^\dagger_{\mathrm{R},\sigma}(x)\Psi^\dagger_{\mathrm{R},\sigma'}(x')U(x,x')\Psi_{\mathrm{R},\sigma'}(x')\Psi_{\mathrm{R},\sigma}(x)}_{\text{Right interaction}} \\
&+\underbrace{\sum_{\sigma,\sigma'}\int dxdx'\,\Psi^\dagger_{\mathrm{L},\sigma}(x)\Psi^\dagger_{\mathrm{R},\sigma'}(x')U(x,x')\Psi_{\mathrm{R},\sigma'}(x')\Psi_{\mathrm{L},\sigma}(x)}_{\text{Direct interaction}} + \underbrace{\sum_{\sigma,\sigma'}\int dxdx'\,\Psi^\dagger_{\mathrm{L},\sigma}(x)\Psi^\dagger_{\mathrm{R},\sigma'}(x')U(x,x')\Psi_{\mathrm{L},\sigma'}(x')\Psi_{\mathrm{R},\sigma}(x)}_{\text{Exchange interaction}} \\
&+\underbrace{\sum_{\sigma}\int dxdx'\,U(x,x')\Big\{\Psi^\dagger_{\mathrm{L},\sigma}(x)\Psi_{\mathrm{R},\sigma}(x)\big[\hat{n}_\mathrm{L}(x')+\hat{n}_\mathrm{R}(x')\big]+\mathrm{H.c.}\Big\}}_{\text{Tunneling}} + O(T^2_{k,k'}),
\end{aligned}
\tag{A1}
$$

where $\hat{n}(x)=\sum_\sigma \Psi^\dagger_\sigma(x)\Psi_\sigma(x)$. The exchange and direct terms don't contribute to tunneling current, i.e. they both commute with the number operator of each side, see Sec. III, and thus will be neglected. The full effective Hamiltonian then reduces to that given by Eq. (22).

Appendix B: Details of the derivation of the tunneling current

Here the details are given for the evaluation of the tunneling current, Eq. (27). Although the evaluation is quite lengthy and tedious, a lot of details are included for completeness.

Defining $\hat{A}_\sigma(x) = T(x)\Psi^\dagger_{\mathrm{L},\sigma}(x)\Psi_{\mathrm{R},\sigma}(x)$ and $\hat{B}_\sigma(x,x') = U(x,x')\Psi^\dagger_{\mathrm{L},\sigma}(x)\Psi_{\mathrm{R},\sigma}(x)\big[\hat{n}_\mathrm{L}(x')+\hat{n}_\mathrm{L}(x')\big]$ for notational convenance. Note that

$$
\begin{aligned}
\langle\hat{A}_\sigma(x)\rangle_{H_0} &= \langle\hat{B}_\sigma(x,x')\rangle_{H_0} = 0\\
\langle\hat{A}_\sigma(x)\hat{A}_{\sigma'}(x')\rangle_{H_0} &= \langle\hat{B}_\sigma(x,x')\hat{B}_{\sigma'}(x'',x''')\rangle_{H_0} = 0
\end{aligned}
$$

and

$$\langle\hat{A}_\sigma(x)\hat{B}_{\sigma'}(x',x'')\rangle_{H_0} = 0.$$

Evaluating the commutator in Eq. (27), the total current can be written as $I=I_1+I_2+I_3$, where

$$
\begin{aligned}
I_1 = 2e\int_{-\infty}^{t} dt'\sum_{\sigma,\sigma'}\int dxdx'\,\mathrm{Re}\Big[&\langle\hat{A}^\dagger_{\sigma'}(x',t')\hat{A}_\sigma(x,t)\rangle\\
&-\langle\hat{A}_{\sigma'}(x',t')\hat{A}^\dagger_\sigma(x,t)\rangle\Big],
\end{aligned}
\tag{B1}
$$

$$
\begin{aligned}
I_2 = 2e\int_{-\infty}^{t} dt'\sum_{\sigma,\sigma'}\int &dx_1dx_2dx_3dx_4\\
&\times\mathrm{Re}\Big[\langle\hat{B}^\dagger_{\sigma'}(x_1,x_2,t')\hat{B}_\sigma(x_3,x_4,t)\rangle\\
&-\langle\hat{B}_{\sigma'}(x_1,x_2,t')\hat{B}^\dagger_\sigma(x_3,x_4,t)\rangle\Big],
\end{aligned}
\tag{B2}
$$

and

$$
\begin{aligned}
&I_3 = 2e\int_{-\infty}^{t} dt'\sum_{\sigma,\sigma'}\int dx'dx_1dx_2\\
&\mathrm{Re}\Big[\langle\hat{A}^\dagger_{\sigma'}(x',t')\hat{B}_\sigma(x_1,x_2,t)\rangle - \langle A_{\sigma'}(x',t')\hat{B}^\dagger_\sigma(x_1,x_2,t)\rangle\\
&+\langle\hat{B}^\dagger_{\sigma'}(x_1,x_2,t')\hat{A}_\sigma(x',t)\rangle - \langle\hat{B}_{\sigma'}(x_1,x_2,t')\hat{A}^\dagger_\sigma(x',t)\rangle\Big].
\end{aligned}
\tag{B3}
$$

The first two contributions will be evaluated in the following sections, while the third term, I_3, will be neglected. As it contains two-particle correlations with three equal times. It would be expected that these correlations are subdominant by this restriction of phase-space.

1. I_1

To evaluate the expectation values in Eq. (B1) it is useful to introduce the Keldysh contour-ordered single-particle Green's function[19]

$$G^X_\sigma(x,t,t') = -i\big\langle T_C\Psi_{X,\sigma}(x,t)\Psi^\dagger_{X,\sigma}(x,t')\big\rangle_{H_X}, \tag{B4}$$

where $X\in\{\mathrm{L},\mathrm{R}\}$. The quantities of interest for this work are the "lesser" ($<$) and "greater" ($>$) functions

given by

$$\left[G_\sigma^X(x,t,t')\right]^< = i\left\langle \Psi^\dagger_{X,\sigma}(x,t')\Psi_{X,\sigma}(x,t)\right\rangle_{H_X} = i\int d\omega\, e^{-i\omega(t-t')} n_{\mathrm{F}}(\omega)\rho_{X,\sigma}(x,\omega) \tag{B5}$$

and

$$\left[G_\sigma^X(x,t,t')\right]^> = -i\left\langle \Psi_{X,\sigma}(x,t)\Psi^\dagger_{X,\sigma}(x,t')\right\rangle_{H_X} = -i\int d\omega\, e^{-i\omega(t-t')}\left[1-n_{\mathrm{F}}(\omega)\right]\rho_{X,\sigma}(x,\omega), \tag{B6}$$

where $n_{\mathrm{F}}(\omega) = (e^{\beta\omega}+1)^{-1}$ is the Fermi distribution with inverse temperature $\beta = (k_{\mathrm{B}}T)^{-1}$ and $\rho^{\mathrm{I}}_\sigma(x,\omega)$ is the single-particle local density of states, obtained from the imaginary part of the retarded Green's function,

$$\rho_\sigma(x,\omega) = -\frac{1}{\pi}\mathrm{Im}\, G^{\mathrm{ret}}_\sigma(x,\omega). \tag{B7}$$

In the limit were the tunneling is spatially localized about a single point, as it for an STM, where the wave functions of the tip are exponentially localized on the atomic scale about the location of the tip, the spatial integrals in (B1) can be approximated by the value of the functions centered at the tunneling point. This is equivalent to the, common, assumption that the tunneling matrix elements $T_{\mathbf{k},\mathbf{k}'} = \langle\psi_{\mathrm{L},\mathbf{k}}|T|\psi_{\mathrm{R},\mathbf{k}'}\rangle$ have no momentum dependance. The needed correlation functions from (B1) are then (assuming spin is conserved during tunneling)

$$\langle \hat{A}^\dagger_\sigma(x,t')\hat{A}_\sigma(x,t)\rangle_{H_0} = |T|^2\langle\Psi_{\mathrm{L},\sigma}(x,t')\Psi^\dagger_{\mathrm{L},\sigma}(x,t)\rangle_{H_{\mathrm{L}}}\langle\Psi^\dagger_{\mathrm{R},\sigma}(x,t')\Psi_{\mathrm{R},\sigma}(x,t)\rangle_{H_{\mathrm{R}}} \tag{B8}$$

and

$$\langle \hat{A}_\sigma(x,t')\hat{A}^\dagger_\sigma(x,t)\rangle_{H_0} = |T|^2\langle\Psi^\dagger_{\mathrm{L},\sigma}(x,t')\Psi_{\mathrm{L},\sigma}(x,t)\rangle_{H_{\mathrm{L}}}\langle\Psi_{\mathrm{R},\sigma}(x,t')\Psi^\dagger_{\mathrm{R},\sigma}(x,t)\rangle_{H_{\mathrm{R}}}. \tag{B9}$$

By using Eqs. (B5) and (B6), the well-known result relating the tunneling current to the single-particle local density of states is recovered,

$$I_1 = 2\pi e|T|^2\sum_\sigma\int d\omega\,\rho_{\mathrm{L},\sigma}(x,\omega+eV)\rho_{\mathrm{R},\sigma}(x,\omega) \times\left[n_{\mathrm{F}}(\omega)-n_{\mathrm{F}}(\omega+eV)\right]. \tag{B10}$$

2. I_2

The remaining contribution to the tunneling current, I_2, contains many high order correlations functions, including three particle propagators. The evaluations of which becomes quite tedious and lengthy. But as this is directly related to this work, we will cover these in some detail.

From Eq. (B2), the needed correlations for I_2 are

$$\begin{aligned}
&\langle \hat{B}^\dagger_\sigma(x_1,x_2,t')\hat{B}_\sigma(x_3,x_4,t)\rangle_{H_0} = \\
&\quad U(x_1,x_2)U(x_3,x_4)\Big[\langle\hat{n}_{\mathrm{L}}(x_2,t')\Psi_{\mathrm{L},\sigma}(x_1,t')\Psi^\dagger_{\mathrm{L},\sigma}(x_3,t)\hat{n}_{\mathrm{L}}(x_4,t)\rangle_{H_{\mathrm{L}}}\langle\Psi^\dagger_{\mathrm{R},\sigma}(x_1,t')\Psi_{\mathrm{R},\sigma}(x_3,t)\rangle_{H_{\mathrm{R}}} \\
&\quad + \langle\Psi_{\mathrm{L},\sigma}(x_1,t')\Psi^\dagger_{\mathrm{L},\sigma}(x_3,t)\rangle_{H_{\mathrm{L}}}\langle\hat{n}_{\mathrm{R}}(x_2,t')\Psi^\dagger_{\mathrm{R},\sigma}(x_1,t')\Psi_{\mathrm{R},\sigma}(x_3,t)\hat{n}_{\mathrm{R}}(x_4,t)\rangle_{H_{\mathrm{R}}} \\
&\quad + \langle\hat{n}_{\mathrm{L}}(x_2,t')\Psi_{\mathrm{L},\sigma}(x_1,t')\Psi^\dagger_{\mathrm{L},\sigma}(x_3,t)\rangle_{H_{\mathrm{L}}}\langle\Psi^\dagger_{\mathrm{R},\sigma}(x_1,t')\Psi_{\mathrm{R},\sigma}(x_3,t)\hat{n}_{\mathrm{R}}(x_4,t)\rangle_{H_{\mathrm{R}}} \\
&\quad + \langle\Psi_{\mathrm{L},\sigma}(x_1,t')\Psi^\dagger_{\mathrm{L},\sigma}(x_3,t)\hat{n}_{\mathrm{L}}(x_4,t)\rangle_{H_{\mathrm{L}}}\langle\hat{n}_{\mathrm{R}}(x_2,t')\Psi^\dagger_{\mathrm{R},\sigma}(x_1,t')\Psi_{\mathrm{R},\sigma}(x_3,t)\rangle_{H_{\mathrm{R}}}\Big],
\end{aligned} \tag{B11}$$

and

$$\begin{aligned}
&\langle \hat{B}_\sigma(x_1,x_2,t')\hat{B}^\dagger_\sigma(x_3,x_4,t)\rangle_{H_0} = \\
&\quad U(x_1,x_2)U(x_3,x_4)\Big[\langle\Psi^\dagger_{\mathrm{L},\sigma}(x_1,t')\hat{n}_{\mathrm{L}}(x_2,t')\hat{n}_{\mathrm{L}}(x_4,t)\Psi_{\mathrm{L},\sigma}(x_3,t)\rangle_{H_{\mathrm{L}}}\langle\Psi_{\mathrm{R},\sigma}(x_1,t')\Psi^\dagger_{\mathrm{R},\sigma}(x_3,t)\rangle_{H_{\mathrm{R}}} \\
&\quad + \langle\Psi^\dagger_{\mathrm{L},\sigma}(x_1,t')\Psi_{\mathrm{L},\sigma}(x_3,t)\rangle_{H_{\mathrm{L}}}\langle\Psi_{\mathrm{R},\sigma}(x_1,t')\hat{n}_{\mathrm{R}}(x_2,t')\hat{n}_{\mathrm{R}}(x_4,t)\Psi^\dagger_{\mathrm{R},\sigma}(x_3,t)\rangle_{H_{\mathrm{R}}} \\
&\quad + \langle\Psi^\dagger_{\mathrm{L},\sigma}(x_1,t')\hat{n}_{\mathrm{L}}(x_2,t')\Psi_{\mathrm{L},\sigma}(x_3,t)\rangle_{H_{\mathrm{L}}}\langle\Psi_{\mathrm{R},\sigma}(x_1,t')\hat{n}_{\mathrm{R}}(x_4,t)\Psi^\dagger_{\mathrm{R},\sigma}(x_3,t)\rangle_{H_{\mathrm{R}}} \\
&\quad + \langle\Psi^\dagger_{\mathrm{L},\sigma}(x_1,t')\hat{n}_{\mathrm{L}}(x_4,t)\Psi_{\mathrm{L},\sigma}(x_3,t)\rangle_{H_{\mathrm{L}}}\langle\Psi_{\mathrm{R},\sigma}(x_1,t')\hat{n}_{\mathrm{R}}(x_2,t')\Psi^\dagger_{\mathrm{R},\sigma}(x_3,t)\rangle_{H_{\mathrm{R}}}\Big].
\end{aligned} \tag{B12}$$

Neglecting all two-particle correlations that have three equal times, as these are expected to be sub-dominate from the reduction of phase space, leads to

$$\langle \hat{B}^\dagger_\sigma(x_1,x_2,t')\hat{B}_\sigma(x_3,x_4,t)\rangle_{H_0} \approx U(x_1,x_2)U(x_3,x_4)\Big[\langle \hat{n}_{\rm L}(x_2,t')\Psi_{{\rm L},\sigma}(x_1,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\hat{n}_{\rm L}(x_4,t)\rangle_{H_{\rm L}}\langle \Psi^\dagger_{{\rm R},\sigma}(x_1,t')\Psi_{{\rm R},\sigma}(x_3,t)\rangle_{H_{\rm R}} + \langle \Psi_{{\rm L},\sigma}(x_1,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\rangle_{H_{\rm L}}\langle \hat{n}_{\rm R}(x_2,t')\Psi^\dagger_{{\rm R},\sigma}(x_1,t')\Psi_{{\rm R},\sigma}(x_3,t)\hat{n}_{\rm R}(x_4,t)\rangle_{H_{\rm R}}\Big] \tag{B13}$$

and

$$\langle \hat{B}_\sigma(x_1,x_2,t')\hat{B}^\dagger_\sigma(x_3,x_4,t)\rangle_{H_0} \approx U(x_1,x_2)U(x_3,x_4)\Big[\langle \Psi^\dagger_{{\rm L},\sigma}(x_1,t')\hat{n}_{\rm L}(x_2,t')\hat{n}_{\rm L}(x_4,t)\Psi_{{\rm L},\sigma}(x_3,t)\rangle_{H_{\rm L}}\langle \Psi_{{\rm R},\sigma}(x_1,t')\Psi^\dagger_{{\rm R},\sigma}(x_3,t)\rangle_{H_{\rm R}} + \langle \Psi^\dagger_{{\rm L},\sigma}(x_1,t')\Psi_{{\rm L},\sigma}(x_3,t)\rangle_{H_{\rm L}}\langle \Psi_{{\rm R},\sigma}(x_1,t')\hat{n}_{\rm R}(x_2,t')\hat{n}_{\rm R}(x_4,t)\Psi^\dagger_{{\rm R},\sigma}(x_3,t)\rangle_{H_{\rm R}}\Big]. \tag{B14}$$

To evaluate the remaining three-particle correlations it will be useful to introduce the following contour-order quantities

$$C^{\rm L}_\sigma(t',t) = \langle T_C \hat{n}_{\rm L}(x_1,t')\Psi_{{\rm L},\sigma}(x_2,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\hat{n}_{\rm L}(x_4,t)\rangle_{H_{\rm L}}, \tag{B15}$$

and

$$C^{\rm R}_\sigma(t',t) = \langle T_C \hat{n}_{\rm L}(x_1,t')\Psi^\dagger_{{\rm R},\sigma}(x_2,t')\Psi_{{\rm R},\sigma}(x_3,t)\hat{n}_{\rm R}(x_4,t)\rangle_{H_{\rm R}}. \tag{B16}$$

Such that

$$\big[C^{\rm L}_\sigma(t',t)\big]^> = \langle \hat{n}_{\rm L}(x_1,t')\Psi_{{\rm L},\sigma}(x_2,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\hat{n}_{\rm L}(x_4,t)\rangle_{H_{\rm L}}, \tag{B17}$$

$$\big[C^{\rm L}_\sigma(t',t)\big]^< = -\langle \Psi^\dagger_{{\rm L},\sigma}(x_3,t)\hat{n}_{\rm L}(x_4,t)\hat{n}_{\rm L}(x_1,t')\Psi_{{\rm L},\sigma}(x_2,t')\rangle_{H_{\rm L}}, \tag{B18}$$

and

$$\big[C^{\rm R}_\sigma(t',t)\big]^> = \langle \hat{n}_{\rm R}(x_1,t')\Psi^\dagger_{{\rm R},\sigma}(x_2,t')\Psi_{{\rm R},\sigma}(x_3,t)\hat{n}_{\rm R}(x_4,t)\rangle_{H_{\rm R}}, \tag{B19}$$

$$\big[C^{\rm R}_\sigma(t',t)\big]^< = -\langle \Psi_{{\rm R},\sigma}(x_3,t)\hat{n}_{\rm R}(x_4,t)\hat{n}_{\rm R}(x_1,t')\Psi^\dagger_{{\rm R},\sigma}(x_2,t')\rangle_{H_{\rm R}}. \tag{B20}$$

The three-particle correlation functions (B15) and (B16) can be expressed as, neglecting two-particle correlations with three equal times and the three-particle vertex,

$$\begin{aligned} C^{\rm L}_\sigma(t',t) \approx & \sum_{\sigma'}\langle T_C\Psi^\dagger_{{\rm L},\sigma'}(x_1,t')\Psi_{{\rm L},\sigma'}(x_4,t)\rangle\langle T_C\Psi_{{\rm L},\sigma'}(x_1,t')\Psi_{{\rm L},\sigma}(x_2,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\Psi^\dagger_{{\rm L},\sigma'}(x_4,t)\rangle \\ & - \sum_{\sigma'}\langle T_C\Psi_{{\rm L},\sigma}(x_1,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\rangle\langle T_C\Psi^\dagger_{{\rm L},\sigma}(x_1,t')\Psi_{{\rm L},\sigma}(x_2,t')\Psi^\dagger_{{\rm L},\sigma'}(x_4,t)\Psi_{{\rm L},\sigma'}(x_4,t)\rangle \\ & + \sum_{\sigma'}\langle T_C\Psi_{{\rm L},\sigma'}(x_1,t')\Psi^\dagger_{{\rm L},\sigma'}(x_4,t)\rangle\langle T_C\Psi^\dagger_{{\rm L},\sigma'}(x_1,t')\Psi_{{\rm L},\sigma}(x_2,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\Psi_{{\rm L},\sigma'}(x_4,t)\rangle \\ & - \sum_{\sigma'}\langle T_C\Psi_{{\rm L},\sigma}(x_2,t')\Psi^\dagger_{{\rm L},\sigma}(x_4,t)\rangle\langle T_C\Psi^\dagger_{{\rm L},\sigma'}(x_1,t')\Psi_{{\rm L},\sigma'}(x_1,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\Psi_{{\rm L},\sigma}(x_4,t)\rangle \\ & + \langle T_C\Psi_{{\rm L},\sigma}(x_2,t')\Psi^\dagger_{{\rm L},\sigma}(x_3,t)\rangle\langle T_C\hat{n}_{\rm L}(x_1,t')\hat{n}_{\rm L}(x_4,t)\rangle. \end{aligned} \tag{B21}$$

A similar expression exists for $C^{\rm R}_\sigma(t',t)$, Eq. (B16).

Again assuming we are in the point contact or localized tunneling regime, where the wave functions of one side, say L, are spatially localized about the tunneling point, x, the dominate contribution from the spatial integrals of (B2) can be approximated by the value at x. This is equivalent to neglecting the momentum dependance of the matrix elements of the interaction $U_{\mathbf{k},\mathbf{k}'} = \langle\psi_{{\rm L},\mathbf{k}}|U|\psi_{{\rm R},\mathbf{k}}\rangle$, which for a good metal is approximately true; especially for the

small energy region about the Fermi energy that one is typically interested in for tunneling experiments. Also for a contour-ordered quantity such as $X(t,t')=Y(t,t')Z(t,t')$, the greater and lesser functions are simply giving by $\big[X(t,t')\big]^{\gtrless}=\big[Y(t,t')\big]^{\gtrless}\big[Z(t,t')\big]^{\gtrless}$. Along with defining

$$\chi^{X}_{\sigma_1,\sigma_2,\sigma_3,\sigma_4}(x,t-t')=\langle\Psi^{\dagger}_{X,\sigma_1}(x,t)\Psi_{X,\sigma_2}(x,t)\Psi^{\dagger}_{X,\sigma_3}(x,t')\Psi_{X,\sigma_4}(x,t')\rangle=\int\frac{d\omega}{2\pi}\,\chi^{X}_{\sigma_1,\sigma_2,\sigma_3,\sigma_4}(x,\omega)e^{-i\omega(t-t')},\tag{B22}$$

and the two-particle density of states $\rho^{\mathrm{II}}_{\sigma,\overline{\sigma}}(x,\omega)=-\frac{1}{\pi}\mathrm{Im}\,G^{\mathrm{II}}_{\sigma,\overline{\sigma}}(x,\omega)$, where

$$G^{\mathrm{II}}_{\sigma,\overline{\sigma}}(x,t)=-i\theta(t)\langle\{\Psi_{\sigma}(x,t)\Psi_{\overline{\sigma}}(x,t),\Psi^{\dagger}_{\overline{\sigma}}(0)\Psi^{\dagger}_{\sigma}(0)\}\rangle_{H},\tag{B23}$$

Eq. (B2) can finally be evaluated and is giving by

$$\begin{aligned}
&I_2=\\
&eU^2\sum_{\sigma,\sigma'}\int_{-\infty}^{\infty}d\omega d\omega'\,\rho_{\mathrm{L},\sigma}(x,\omega+eV)\rho_{\mathrm{R},\sigma'}(x,\omega')\Big\{\big[\chi^{\mathrm{L}}_{\overline{\sigma}',\overline{\sigma},\overline{\sigma},\overline{\sigma}'}(x,\omega'-\omega-eV)+\chi^{\mathrm{R}}_{\overline{\sigma}',\overline{\sigma},\overline{\sigma},\overline{\sigma}'}(x,\omega'-\omega)\big]n_{\mathrm{F}}(\omega')\big[1-n_{\mathrm{F}}(\omega+eV)\big]\\
&-\big[\chi^{\mathrm{L}}_{\overline{\sigma},\overline{\sigma}',\overline{\sigma}',\overline{\sigma}}(x,\omega-\omega'+eV)+\chi^{\mathrm{R}}_{\overline{\sigma},\overline{\sigma}',\overline{\sigma}',\overline{\sigma}}(x,\omega-\omega')\big]n_{\mathrm{F}}(\omega+eV)\big[1-n_{\mathrm{F}}(\omega')\big]\Big\}\\
&+e\,\pi U^2\sum_{\sigma}\int_{-\infty}^{\infty}d\omega d\omega'\,\rho_{\mathrm{L},\overline{\sigma}}(x,\omega+eV)\rho_{\mathrm{R},\sigma}(x,\omega')\rho^{\mathrm{II}}_{\mathrm{L},\sigma,\overline{\sigma}}(x,\omega+\omega'+eV)\\
&\times\Big\{n_{\mathrm{F}}(\omega+eV)n_{\mathrm{F}}(\omega')\big[1-n_{\mathrm{F}}(\omega+\omega'+eV)\big]-\big[1-n_{\mathrm{F}}(\omega+eV)\big]\big[1-n_{\mathrm{F}}(\omega')\big]n_{\mathrm{F}}(\omega+\omega'+eV)\Big\}\\
&-e\,\pi U^2\sum_{\sigma}\int_{-\infty}^{\infty}d\omega d\omega'\,\rho_{\mathrm{L},\overline{\sigma}}(x,\omega+eV)\rho_{\mathrm{R},\sigma}(x,\omega')\rho^{\mathrm{II}}_{\mathrm{R},\sigma,\overline{\sigma}}(x,\omega+\omega')\\
&\times\Big\{n_{\mathrm{F}}(\omega+eV)n_{\mathrm{F}}(\omega')\big[1-n_{\mathrm{F}}(\omega+\omega')\big]-\big[1-n_{\mathrm{F}}(\omega+eV)\big]\big[1-n_{\mathrm{F}}(\omega')\big]n_{\mathrm{F}}(\omega+\omega')\Big\},
\end{aligned}\tag{B24}$$

which is Eq. (31).

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